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Neutron Radiographic Investigations of Hydride Blisters Grown on Zr-2.5Nb Pressure Tube Spool Piece Under Stimulated Condition of In-Reactor Pressure and Temperature

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Abstract

The present work has been carried out with an aim to have a thorough understanding of the mechanism of blister formation and the resulting degradation in the serviceability of the component under simulated in-reactor condition of pressure and temperature. A 220 MWe Indian PHWR Zr-2.5Nb pressure tube spool piece of 165 mm length was charged with a homogeneous hydrogen concentration using lithium hydroxide solution at 300 °C followed by growth of multiple hydride blisters at 10 different locations on the pressure tube outside surface by maintaining cold spots. This blistered tube was subjected to burst test by pressurizing the tube in steps of 20 kg/cm^2 for 10 minutes up to 380 kg/cm^2 at which it burst with sound. The failed tube was examined by neutron radiography, scanning electron microscopy, surface profiling, video microscopy and digital imaging for characterizing the cracked and un-cracked blisters. Neutron radiographs of the hydride blisters were recorded using NR facility at Apsara, a 200 kW swimming pool type reactor using transfer technique with 25 μm Gd neutron converter screen. The details of NR investigations done are reported in this paper.

Keywords: *Zr-2.5Nb, Pressure tube, Hydride blisters, Neutron radiography*

1. Introduction

Pressure tubes are the most vulnerable zirconium alloy components in the Pressurised Heavy Water Reactor (PHWR) core. The pressure tubes operate in severe environment of high temperature (~ 300°C), high pressure (112 kg/cm^2) and high neutron flux ($3x10^{13}$ $n/cm^2/sec$). The pressure tubes are surrounded the calandria tubes operating at 70°C. A simplified sketch of a PHWR fuel channel is shown in Fig.1. The new generation of Indian Pressurised Heavy Water Reactors (PHWRs) has Zr-2.5Nb alloy pressure tubes in cold worked and stress relieved condition. Although the hydrogen content of the pressure tube is kept as low as possible (~ 5 ppm), it can pick up hydrogen/deuterium during its in-reactor service life [1]. Hydrogen can cause severe deterioration in the structural integrity and eventually failure of zirconium pressure tubes through gross hydride embrittlement due reorientation of hydride plates under stress and localized embrittlement [2]. The gross embrittlement requires a certain minimum volume fraction of the hydride phase to be present. In case of localized embrittlement, hydrogen absorbed in the tube can migrate to the cold spot formed due to contact between the pressure tube and the calendria tube under the action of thermal gradient and form a hydride blister (Fig.2). These blisters hard and brittle could eventually grow and crack. Formation of series of cracked blisters could lead to rupture of the pressure tube by the delayed hydrogen cracking mechanism.

Studies on the mechanism of hydrogen-induced damages in pressure tubes of Zr based alloys have been done on small coupons cut from pressure tube due to either cost of the experiment or technical difficulties involved. Extrapolating these results on the actual component always has some factor of uncertainties involved due to the size differences between the specimen and component. The present work has been carried out with an aim to have a thorough understanding of the mechanism of blister formation and the resulting degradation in the serviceability of the component under simulated in-reactor condition of pressure and temperature. Various techniques like neutron radiography, scanning electron microscopy, surface profiling, video microscopy and digital imaging have been used for characterizing the cracked and un-cracked blisters. The details of NR investigations done are reported in this paper.

2. Experimental

2.1 Generation of Hydride Blisters

Various methods of charging hydrogen in zirconium alloys have been reported in the literature. These include high temperature cathodic charging, corrosion in lithium hydroxide solution in an autoclave and gas hydriding. A 220 MWe Indian PHWR Zr-2.5Nb pressure tube spool piece of 165 mm length was charged with a homogeneous hydrogen concentration using lithium hydroxide solution at 300 °C. It was followed by growth of multiple hydride blisters at 10 different locations on the outside surface of the pressure tube by maintaining cold spots. Water cooled hollow brass cones of different tip diameters (3 mm- 8 mm) were used for simulation of cold spots with temperature of 30 °C - 40°C. Diffusion of hydrogen from hot regions to cold spots resulted in zirconium hydride blisters at these spots. The blisters were generated at positions located symmetrically with respect to tube ends in two groups (A and B) of five each (1-5). The axial separation between the groups was 50 mm. Within the group, the blisters were located at different angular positions. The schematic of the distribution of blisters on the outside surface of the pressure tube with their identification is shown in Fig. 3. This blistered tube was subjected to burst test by pressurizing the tube in steps of 20 kg/cm2 for 10 minutes up to 380 kg/cm2 at which it burst with sound. The details of experiment of growing blisters and burst test of the blistered tube are published elsewhere [3].

2.2 Neutron Radiography

The property of thermal neutrons, which makes them valuable for studying industrial components, is their high penetration through widely used industrial materials such as steel, aluminium or zirconium. Neutrons are efficiently attenuated by only a few specific elements such as hydrogen, boron, cadmium, samarium and gadolinium. For example, organic materials or water attenuate neutrons because of their high hydrogen content, while many structural materials such as aluminium or steel are nearly transparent. Neutron Radiography (NR) is a non-destructive imaging technique similar to X-ray and gamma radiography for material testing and is based on the local variations in absorption encountered by a beam in simple transmission [4]. It has some special advantages in nuclear, aerospace, ordnance and rubber & plastic industries. It is very valuable in practice because most of the materials have very low absorption for thermal neutrons, and therefore it is possible to investigate very bulky objects and selectively see those parts with high real or apparent absorption cross-section. A converter material in the form of a foil or film/plate coated with neutron absorbing material that emits light or charged particles is used in Neutron Radiography.

Neutron Radiographic Investigations

Figure 1 shows the basic principle of radiography methods. The object under examination is placed in the path of a well collimated beam of neutrons from either a reactor or a non-reactor source and the transmitted beam is detected. During transmission, the incident beam undergoes intensity modulation due to the absorption and scattering properties of the material. Any inhomogeneity in the object such as internal structure, defects like voids, cracks, inclusions and porosity will be shown up as a change in the detected intensity recorded behind the object as shown in the Fig.1. The neutron beam transmitted through the object is recorded using a suitable imaging method.

Neutron radiography is capable of detecting inclusions containing low atomic number elements in a matrix of relatively higher atomic number materials. Presence of zirconium hydride blisters/platelets in zirconium matrix can therefore be detected by neutron radiography. A programme of examining hydride blisters created in the laboratory on zircaloy/ Zr-2.5Nb pressure tubes was undertaken using neutron radiography [5,9]. It was observed that a zirconium hydride blister of nearly 0.35% of job thickness could be detected using neutron radiography. This work served as a reference in examining irradiated zircaloy pressure tubes from a power reactor with residence time of ~ 8.25 effective full power years and which had developed hydride blisters. Neutron radiography was also used to study size and shape of the zirconium hydride blisters embedded in the zircaloy-2 pressure tube.

Neutron radiography was done at Apsara, a 400 kw swimming pool type reactor using thermal neutrons collimated by a divergent collimator with L/D ratio of 90. Radiographs were taken by direct technique using 25μm Gd screen and D-7 type industrial x-ray film. A schematic diagram of NR facility at Apsara is shown in Fig.5 and details are given in Table 1.

Table 1: Apsara Reactor NR Facility

Useful beam area: 15cm dia
Thermal neutron flux: 1 X 10^6 n/cm^2/sec
Gamma radiation level: 4R/h
Cadmium ratio: 6.3
Neutron /gamma ratio: 9 X 10^5 n/cm^2/mR
Neutron converter screens: Gd 25μ, Dy 100μ, Kodak CN85-B converter screens, ^{6}LiF-ZnS(Ag) scintillator,CCD camera, Fuji neutron image plate.

3. Results and Discussion

The pressure tube was examined and hydride blisters were detected at all the ten cold spot locations. Each location single blister had formed but in case of position 2A, a few small blisters had grown along with the bigger size blister. The blisters were formed in various shapes and sizes depending on the size of the cold tips touching the surface of the pressure tube. Most of the blisters were of elliptical in shape except 1B, 2B, 3B (irregular) and 4B(circular). The unique feature of the pressure tube bursting was that a crack is extending from one end of the tube to the other passed through the middle of two blisters, namely 4A and 4B falling in one line. Fig. 6 shows the digital photographs of (a) some of the blisters on the tube and (b) crack passing through blisters 4A and 4B. After the burst test some of the blisters have been observed to have their edge or the central part broken. A few of them also have been found to have developed cracks. Images of the hydride blisters recorded using video microscope are shown in Fig. 7.

Figure 8 shows neutron radiograph of the pressure tube. All the 10 blisters are clearly seen in the radiograph. The blisters 3A, 3B and 4A, 4B are in the front side and blisters 1A, 1B are seen in the back of the blisters

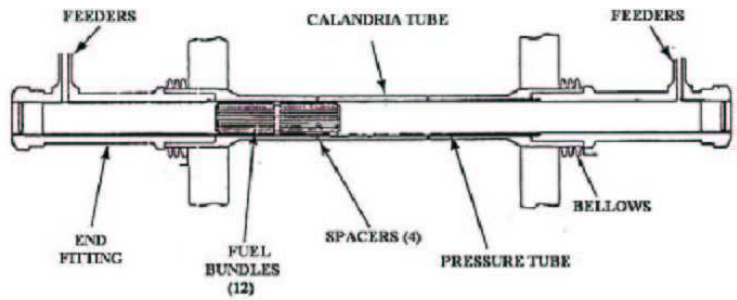


Fig.1: Simplified illustration of a PHWR fuel channel

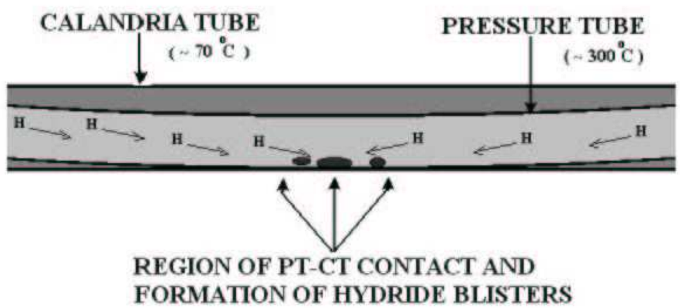


Fig.2: Mechanism of formation of hydride blisters at PT-CT contact in PHWR fuel channel

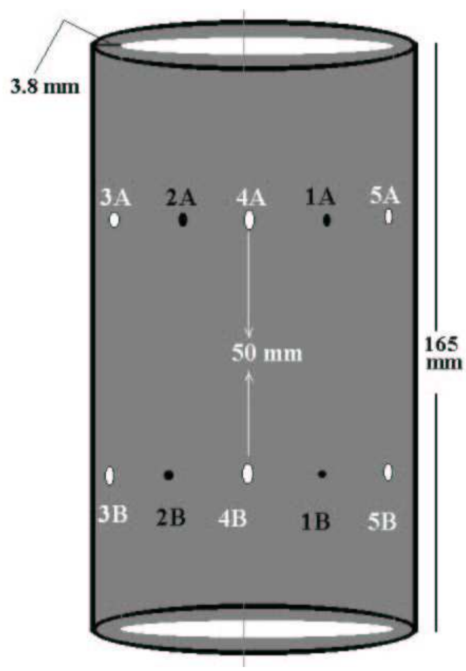


Fig. 3: Schematic of the blister locations on the PT spool piece. Distribution of hydride blisters symmetrically located with respect to tube ends. The blisters 1A, 1B and 2A, 2B (shown in black) are on back side while others (shown in white) are on front side of the tube

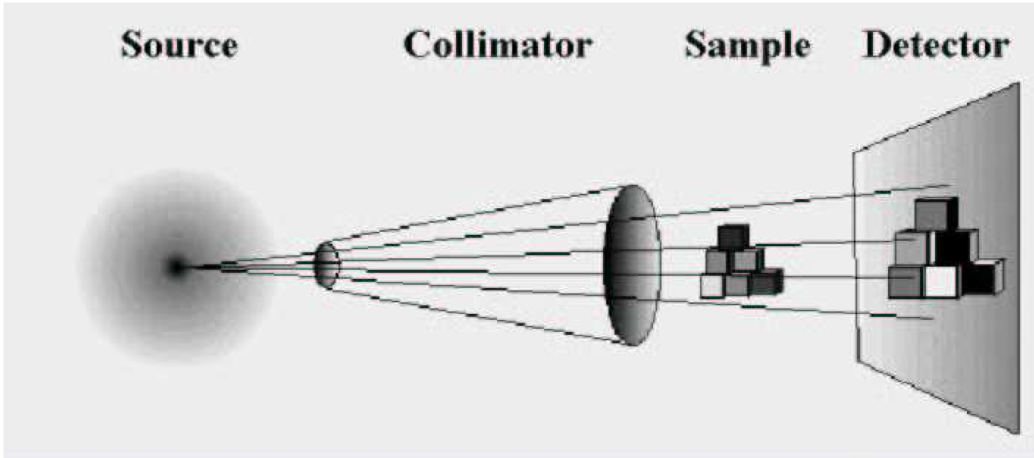


Fig.4: Principle of Neutron Radiography

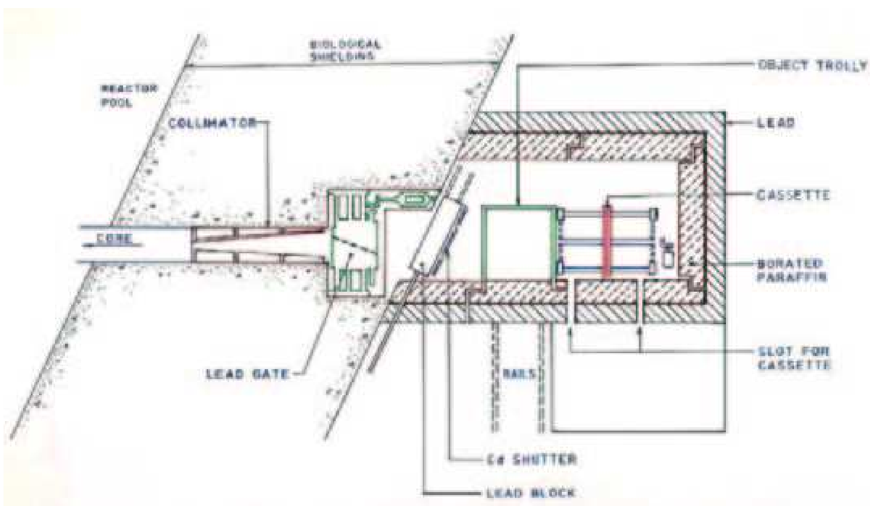
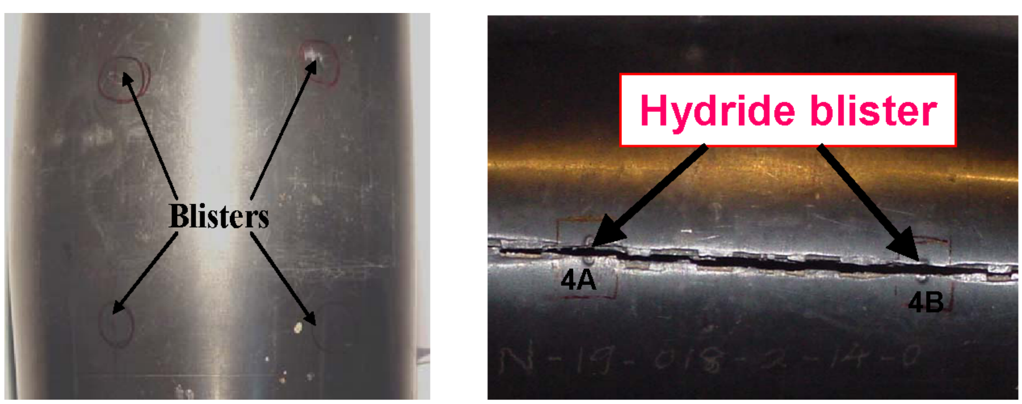


Fig.5: Schematic diagram of NR Facility at Apsara Reactor



(a) (b)

Fig.6: a) Four hydride blisters on one side view of the pressure tube. b) Crack passing through two blisters 4A and 4B

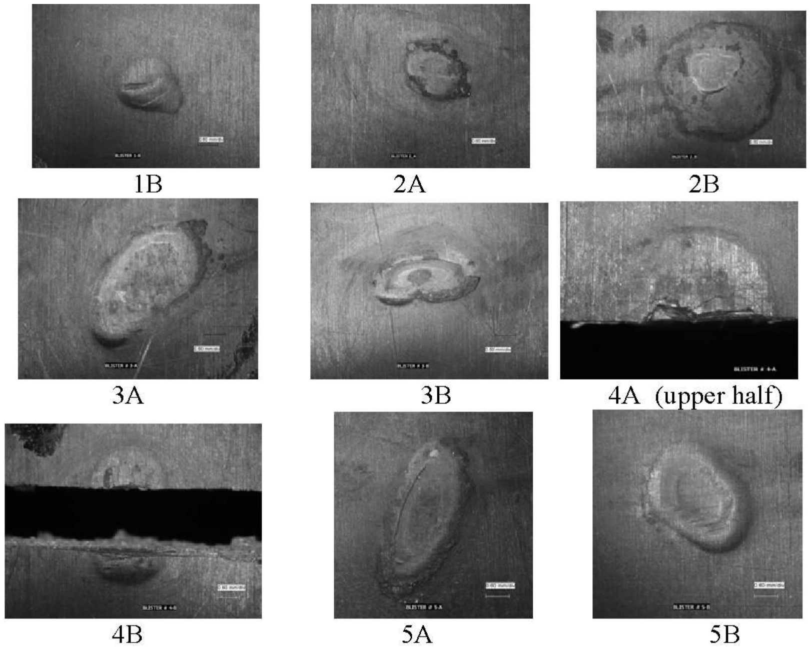


Fig.7: Images of the hydride blisters recorded using video microscope. The scale shown is 0.60 mm/div

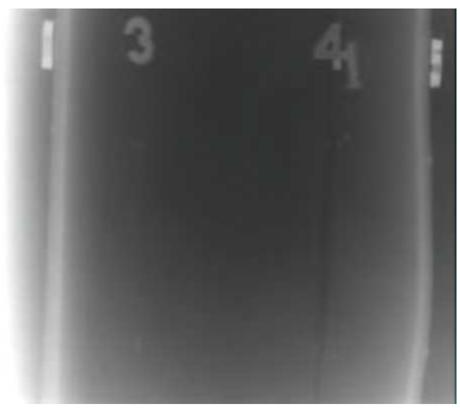


Fig.8: Neutron Radiograph of the pressure tube showing all the 10 blisters and crack passing through 4A and 4B

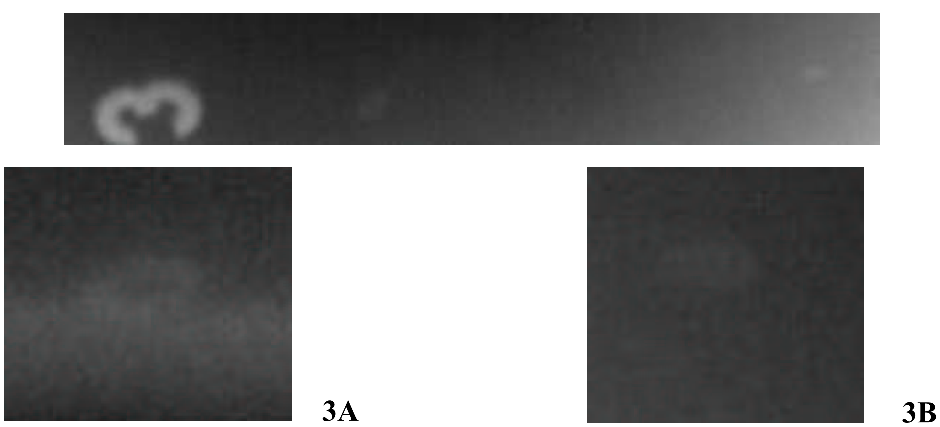


Fig. 9: Neutron Radiograph of blisters 3A and 3B and their enlarged view

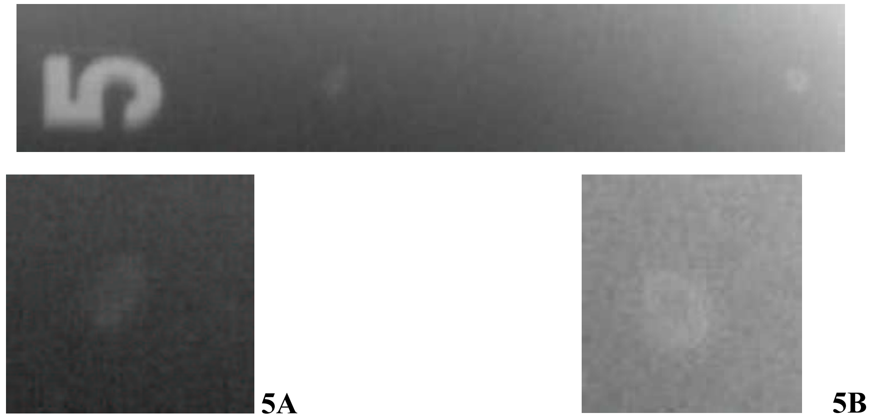


Fig. 10: Neutron radiograph of blister 5A and 5B with their enlarged view

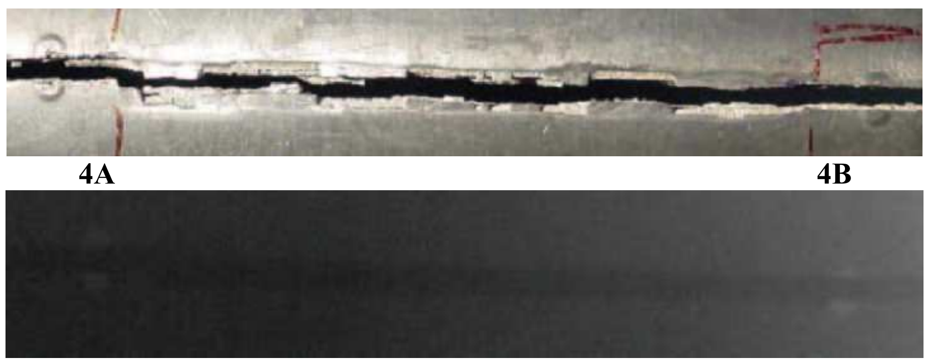
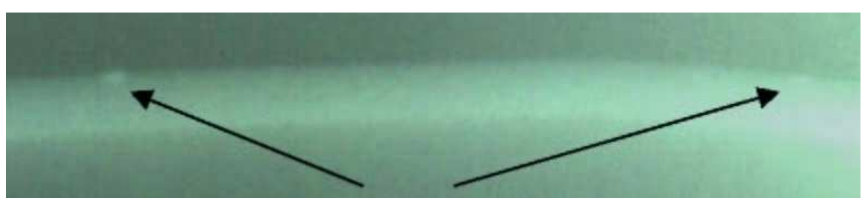
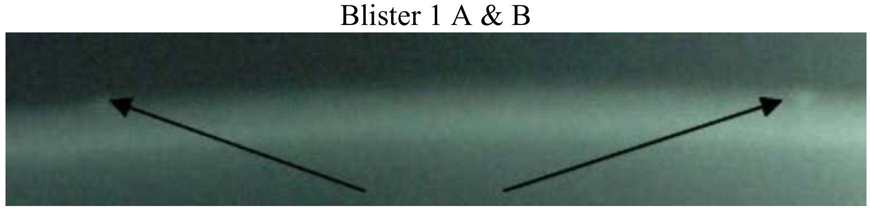


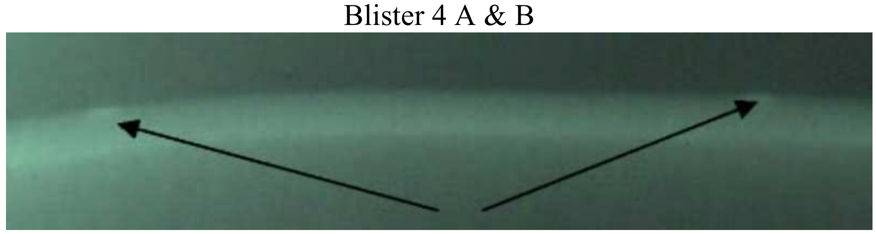
Fig. 11: Photograph of the crack through blisters 4A and 4B in the burst pressure tube (top) and its neutron radiograph (bottom)



Blister 1 A & B



Blister 4 A & B



Blister 5 A & B

Fig. 12 Neutron radiographs of various hydrode blisters with neutron incident parallel to the plane of the blisters

4A and 4B. The blisters 2A, 2B and 5A, 5B are at the two edges of the pressure tube. The radiograph also shows the burst crack passing through middle of the blisters 4A and 4B and extending the complete length of the pressure tube.

Table 2: Sizes of the blisters measured from the neutron radiographs

Blister No.	Shape of the blister	Dimensions in mm Long	Short
1A	Elliptical	2.8	2.3
1B	Irregular	2.4	2.0
2A	Elliptical approx.	3.0	2.3
2B	irregular	2.1	1.8
3A	Elliptical approx.	4.8	2.6
3B	Elliptical approx.	3.4	1.5
4A	Elliptical	3.6	2.2
4B	Circular	2.2	2.1
5A	Elliptical	4.5	2.0
5B	Elliptical	3.4	2.5

Figures 9 to 11 show neutron radiographs of some of the blisters along with their enlarged view. The radiographs reveal information about size, shape and profile of the blisters, which is well compared with that obtained from video microscopy, 3-D surface profiling and other techniques. For example the variation in film optical density in the centre of the 3A and 5A blisters indicate that the central part is broken in the burst test. The shape and sizes of the hydride blisters were determined from neutron radiographs. The details are given in table 2.

Figure 12 shows neutron radiographs of various hydride blisters with neutron beam incident parallel to the plane of the blisters. Lenticular shape of the blister with nearly 2/3 of the blister embedded in the wall of the tube is clearly seen. The average height of the blister above the pressure tube surface is found to be ~ 50- 60 μm. This observation is in agreement with the surface profile measurements [3]. The 4A blister is found to have maximum penetration, more than 700 μm in the pressure tube wall.

4. Conclusion

A 220 MWe Indian PHWR Zr-2.5Nb pressure tube spool piece of 165 mm length was charged with a homogeneous hydrogen concentration using lithium hydroxide solution at 300 °C followed by growth of multiple hydride blisters at 10 different locations on the pressure tube outside surface by maintaining cold spots. This blistered tube was subjected to burst test by pressurizing the tube in steps of 20 kg/cm2 for 10 minutes up to 380 kg/cm2 at which it burst with sound. The failed tube was examined by neutron radiography, using NR facility at Apsara, a 200 kW swimming pool type reactor using transfer technique with 25 μm Gd neutron converter screen. All the 10 blisters in the pressure tubes were detected. The shape and sizes of the hydride blisters and depth of penetration were determined from the radiographs. It is observed that the blister 4A with have maximum depth of penetration, ~ 700 μm has cracked at an internal pressure of 380 Kg/cm2.

5. References

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